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Oxyfuel combustion in a bubbling fluidized bed combustor

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Abstract

This paper deals with characterization of the oxyfuel combustion in a bubbling fluidized bed. The paper proposes a calculation procedure to evaluate gas flow streams, adiabatic flame temperature and concentrations of species in the flue gas. The second part describes a laboratory scale 30 kW oxyfuel BFB and presents results of experiments with various semi-oxyfuel and full oxyfuel modes. The results are compared with experimental results, the agreement is satisfactory with less than 10 % relative difference. Sustaining of similar fluidization regime along all different cases resulted in decrease of the adiabatic flame temperature that had to be compensated by thermal volumetric load increase from 3.3 to 4.1 MW/m^3.

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Keywords: bubbling fluidized bed; oxyfuel combustion; calculation model; gas streams; adiabatic temperature

1. Introduction

The oxyfuel combustion in a bubbling fluidized bed is only rarely discussed, since CFBs are more favored in current research and development. Nevertheless, it is important to follow up the research in the oxyBFB for its possible application in medium scale plants. The oxyfuel BFB combustion has several important features. Firstly, the sufficient flue gas recirculation must be kept in order to sustain the fluidization. The oxyfuel combustion produces approximately only 20 % of the wet flue gas compared to air combustion. Secondly, properties of the fluidization medium are significantly changed in oxyfuel mode, because the fluidization medium is formed by recirculated flue gas and oxygen. Changed material properties therefore have significant impact on the fluidization regime. Roughly, the minimum

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fluidization velocity can increase by approx. 8.5 % and terminal particle velocity by 4-6 % in oxyfuel mode. Last but not least, in oxyfuel mode is not possible to reach similar thermal and flow conditions as in air-fired mode, as has already been indicated by [1] for a CFB case. A former work [2] studied two oxyBFB cases in comparison with air mode, where the first one was aimed on sustaining similar temperature in the bed. Based on this requirement, the flue gas recycle flow was calculated and the resulting flow of the fluidization medium was about 15-20 % lower compared to the air-fired mode. In the second case, a similar flow of fluidization medium was considered, but in this case a too high drop of the bed temperature of about 150-200°C was calculated. Further details concerning bed temperature and flue gas recirculation supported by experimental results are presented in this paper.

Nomenclature

A_{bed}	cross-section area of the bed zone [m^2]
C	correction coefficient for water vapor condensation in FGR [m^3_N/h]
I_{AD}	specific flue gas enthalpy at adiabatic conditions [kJ/kg]
LHV	lower heating value [kJ/kg]
r	FGR ratio [-]
u_f	fluidization velocity [m/s]
V_A	volume flow of air at normal conditions [m^3_N/h]
$V_{H2O,\,FG}$	volume flow of water vapor in flue gas at normal conditions [m^3_N/h]
$V_{H2O,\,O}$	volume flow of water vapor in oxidizer mixture at normal conditions [m^3_N/h]
$V_{H2O,\,rec}$	volume flow of water vapor in FGR at normal conditions [m^3_N/h]
$V_{recFG,W}$	volume flow of FGR stream, wet, at normal conditions [m^3_N/h]
$V_{FG,D}$	volume flow of flue gas, dry, at normal conditions [m^3_N/h]
$V_{FG,W}$	volume flow of flue gas, wet, at normal conditions [m^3_N/h]
V_{O2}	volume flow of oxygen, at normal conditions [m^3_N/h]
Q_A	specific air sensible heat [kJ/kg]
Q_{fuel}	fuel sensible heat [kJ/kg]
Q_{rec}	specific FGR sensible heat [kJ/kg]
ω''_{H2O}	saturated water vapor volume fraction at given temperature [-]
Z_{CO}	specific loss by unburned CO [-]
Z_C	specific loss by unburned carbon [-]
Z_{solid}	specific loss by sensible heat of outgoing solids [-]

2. Theoretical considerations

Calculation of gas streams in oxyfuel combustion is based on commonly used balance equations assuming ideal combustion, as described in [1], with some specifications to the oxyfuel mode, as discussed e.g. in [2]. The calculations are relative, which means that the outputs are related to a unit of mass of the used fuel. Basic inputs for the calculations are fuel composition and pre-definition of desired oxygen concentration in the fluidization medium. A specific issue is the question of the temperature level of the recirculated flue gas (FGR). In a real application, the FGR stream would be extracted as a clean gas stream, i.e. after the electrostatic precipitator (ESP) or a baghouse filter [3]. The “dirty” FGR extracted before an ESP can be excluded beforehand. After the ESP, the flue gas temperature is typically about 150-200°C. The question is whether to maintain the temperature at the ESP outlet level and return the flue gas back to the air distributor at elevated temperature or whether to cool down the FGR stream. In the first case (so-called wet FGR) the fluidization fan is the most critical point that must sustain the high temperature of the gas stream. The FGR line must be thermally insulated to prevent unwanted condensation. In the second case (so-called dry FGR) the water contained in the flue gas stream is condensed on purpose and a heat exchanger is therefore required. In this case the volumetric flow of the FGR reduces accordingly to its actual temperature, determined by the water vapor saturation pressure.

The gas streams are significantly different in oxyfuel mode. In figure 1 a schematic comparison of gas flows between oxyfuel and air mode is shown. The streams are shown for the same size of a BFB combustor in real scales. The scheme ensures the same flow of fluidization medium in oxyfuel mode as in air mode.

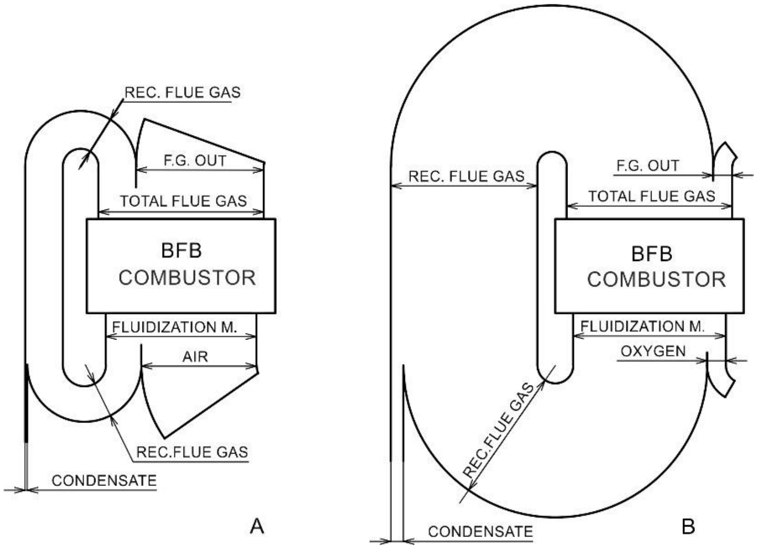

Fig. 1. Volumetric flows (normal thermodynamic conditions) for air (A-left) and oxyfuel (B-right) modes

The air mode also includes a certain part of flue gas recirculation as a measure used to control the bed temperature in the BFB. The stream "condensate" corresponds to cooling the recirculated flue gas down to 40°C in both cases.

Special attention must be paid to considering wet or dry FGR. The flow of the FGR stream in the calculations is introduced through the ratio defined by the equation:

$$r = \frac{V_{recFG,W}}{V_{FG,W}} \tag{1}$$

By this definition, the value of "r" can be higher than 1, which is typically the case for oxyfuel mode. The "r" greater than 1 means that flow of the recirculated flue gas is higher than flow of the flue gas leaving the combustor out to the stack. Without condensation (wet FGR) the amount of water vapor in the recirculated flue gas can be obtained as follows:

$$V_{H_2O,rec} = r.V_{H_2O,FG} \tag{2}$$

The value of $V_{H2O,\,FG}$ is obtained from calculating the amount of water released from combustion of hydrogen and vaporization of water in the fuel. In the case of dry FGR, condensation must be considered. . When using the value of maximal water vapor concentration at a given temperature (equal to saturation pressure), a reduction coefficient "C" respecting the water condensation can be introduced by the equation 3. The resulting volume of the flue gas stream is obtained from calculating the amount of water released from combustion of hydrogen and vaporization of water in the fuel and subtracting the coefficient C.

$$C = \frac{r.\left(V_{H_2O,FG} + V_{H_2O,O} - \frac{\omega''_{H_2O}.V_{FG,D}}{1-\omega''_{H_2O}} \right)}{1+r} \tag{3}$$

Coefficient C is a simplified method for evaluation of the flue gas condensation, respecting the partial pressure of water vapor only. It does not respect the effect of sulfuric acid dew point, which causes an increase of the flue gas dew point temperature. Sulfuric acid is formed by the reaction of sulfur trioxide with water vapor. If we consider the generally accepted fact, that approx. 1 % of all SO_2 in air fired mode to 5 % in oxyfuel mode [4] is converted into SO_3, the maximum concentration of SO_3 is in tens to hundreds of ppm (for reference fuel in Table 1). This amount is negligible in comparison with the concentration of water vapor, which is from about 15 % in air mode to 40 % in oxyfuel mode. This fact allows us to presume that the effect of the sulfuric acid is for the calculation of the coefficient C and for the total volume balance negligible.

The oxyfuel mode also affects the adiabatic flame temperature (T-AD), from which the bed temperature is derived. One of the factors affecting T-AD is a different composition of flue gas and oxidizer, which generally have a higher specific heat capacity in the oxyfuel mode. The specific heat capacity of CO_2 is about 1.6 times higher than the one of N_2 at 1000°C. The second factor is the FGR that is required to maintain the fluidization velocity when the fluidization air flow is decreased (or completely stopped). Without FGR, T-AD would exceed 3000°C in a full oxyfuel mode due to the decreased volume of flue gas that has to carry about the same specific amount of heat as in the air mode. T-AD is obtained from calculation of total heat released in the bed:

$$I_{AD} = LHV.(1 - Z_{CO} - Z_C - Z_{solid}) + Q_{fuel} + Q_{rec} + Q_A \tag{4}$$

Equation 4 equals the specific flue gas enthalpy (obtained from flue gas composition) to the total heat released in the bed. The heat sources are fuel input in form of its lower calorific value, specific heat of the fuel, specific heat carried by the FGR and air (if applicable). The fuel input is reduced by Z_i coefficients that represent losses by incomplete combustion of the fuel (either combustion to CO or unburned) and sensible heat of solids removed from the combustor. Since the specific flue gas enthalpy is temperature dependent, T-AD is obtained by iteration from I_{AD}.

3. Measurement apparatus and calculation inputs

3.1. Oxyfuel BFB combustor

Experimental validation of the calculation procedure was carried out using a 30 kW laboratory scale oxyfuel BFB combustor. A scheme of the combustor is presented in figure 2.

The BFB combustor is modular, consisting of several exchangeable and demountable sections – windbox, distributor, dense bed zone, transitional section with cross section enlargement and freeboard. All sections are heat insulated. Generally, the combustor is divided into combustion zone and freeboard sections, with a freeboard cross-section that is about 2.6x larger compared to the dense bed zone. Primary air is supplied from an air fan, an orifice is used to measure the air flow rate. A similar approach is used for flue gas recirculation (FGR). The FGR is extracted after the cyclone, passes a water-cooled heat exchanger and is introduced shortly before the windbox. The FGR can be considered as semi-dry, since condensation is not complete. Normally, temperature of the FGR after the cooler is about 50-60 °C, while the dew point temperature is about 75°C. The FGR fan takes over the fluidization in oxyfuel mode. Oxygen is supplied from a bottle bundle and is introduced into the FGR tube. The amount of supplied oxygen is measured by a rotameter. The air distributor is made of a perforated metal plate and is exchangeable. The bed height is automatically controlled by using a spillway at 25 cm above the distributor. Excess of bed material is collected in a separate closed vessel that is connected to the spillway. Fuel is supplied from a hopper in a way that it is introduced at the top boundary of the dense bed.

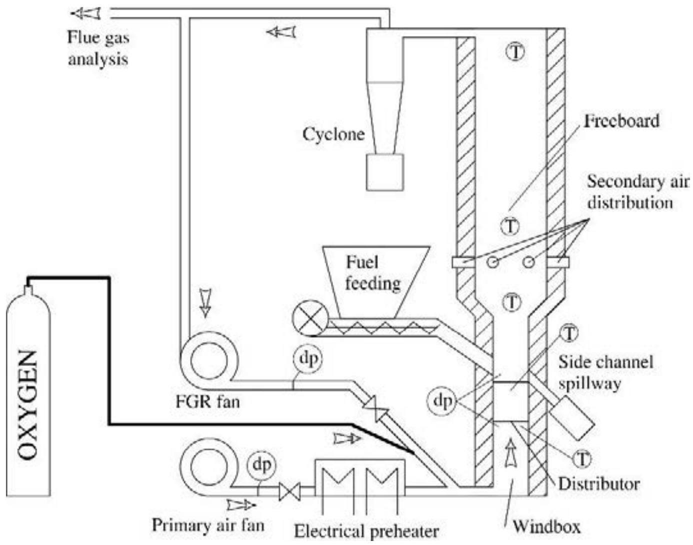

Fig. 2. Scheme of the oxyfuel BFB combustor

Fuel feeding is provided by a screw feeder which is always re-calibrated for a specific fuel. Secondary air input is available at the beginning of the freeboard section, but is not used in the oxyfuel operation. The electrical heater is used in a startup phase and is switched off under normal operation.

Measured values consist of all volume flows of the gas streams (air, oxygen, FGR), mass flow of fuel, bed pressure drop, temperatures and flue gas analysis. The temperature measurement consists of vertical profile in the bed (4 points by 10 cm) and the freeboard (3 points by 45 cm) and temperatures of all gas streams. A mean value of the temperature profile of the bed is referred as the “bed temperature”. The flue gas analysis consists of a standard measurement of O_2, CO, CO_2, NO_x and SO_2 concentrations.

3.2. Fuel specification and inputs

The specification of the fuel that is taken as an input to the calculation model is shown in table 1. The fuel is a high grade lignite type coal having a particle size range 0 – 9 mm that was pre-milled and sieved to limit the upper boundary of the size range. Mean particle diameter is 1.5 mm. The bed material is inherent ash of the coal and its mean particle size is 0.37 mm. Detailed fluidization properties are shown in table 2.

Table 1. Specification of the fuel – lignite coal, as received

C (wt. %)	H (wt. %)	N (wt. %)	S (wt. %)	A (wt. %)	W (wt. %)	LHV (MJ/kg)
47.9	3.9	0.6	0.8	13	18	21.14

Table 2. Properties of the bed material; fluidization velocities calculated on air

density (kg/m^3)	bulk density (kg/m^3)	$d_{p,mean}$ (mm)	minimum fluidization velocity (m/s, at 20/900°C)	terminal velocity (m/s, at 20/900°C)	sphericity (-)
2195	787	0.37	0.37/0.18	2.11/1.91	0.75

The proposed calculation procedure is firstly based on the fact that the measured concentration of oxygen in the flue gas should be the same as the output from calculation. Thus, the calculation is always fixed to a measured parameter. In another way, when a calculation is performed stand-alone, it is decided how much oxygen excess in the flue gas is wanted. After the calculation procedure is performed, the calculated oxygen concentration is obtained back and can be compared with the measured value. The CO_2 concentration is obtained from the calculation as a result and can be compared with the measured CO_2 concentration. Secondly, the next input from measurement is the flow rate of the FGR. The FGR does not affect any concentration in the calculation, therefore it can be used as a variable input. In a stand-alone calculation without experimental data, the flow rate of the FGR should be considered with respect to the requirement to provide a sufficient flow of the fluidization medium. The FGR flow together with power input (defined by fuel feeding rate) also determines the adiabatic flame temperature and consequently the bed temperature. It will be shown in the experimental results that all these parameters vary from case to case, but in combination finally result in the same bed temperature.

To summarize the calculation structure for these specific cases with experimental support, the FGR flow in the calculation is fixed by its measurement in the rig. The oxygen flow is also fixed by its selection at different cases. The last free variable remains the flow of the primary air that is calculated according to the required oxygen concentration in the flue gas, which is determined by measurement.

Even if the balance fuel combustion calculations are normally done as related to a mass unit of fuel burned, in this case they are performed in volume flows. All flows are referred to 273 K and 101.325 kPa. The volume flows are essential in order to evaluate the superficial fluidization velocity. This velocity is obtained from equation 5.

$$u_f = \frac{V_{O_2} + V_A + V_{rec,W}}{A_{bed}} \tag{5}$$

All volume flows must be recalculated to appropriate velocity in order to compare the fluidization velocity with measurement. It must be also noted that the calculation includes water condensation in the FGR. On the basis of the FGR stream composition and measured FGR temperature, the correction factor "C" according to the equation 3 is obtained. T-AD is obtained from equation 4, using the volumetric calculations, database available data on specific enthalpy of each component and using flue gas composition and temperature measurement for specification of Z_i.

4. Results and discussion

The experimental program consisted of several runs with stable operating conditions under different semi-oxyfuel modes, one full oxyfuel mode and one air-only mode with FGR. The aim was to maintain the bed temperature in all cases at 890°C (± 10°C) and the target oxygen concentration in the flue gas at 3 – 5 vol. %. For the oxyfuel case, the concentration of oxygen in the primary oxidizing stream was maintained at 15-17 %. The semi-oxyfuel modes were important to verify the calculation procedure in these transition cases. The first fixed point in the experiments was an oxygen flow that was set to a certain value in each case. The second fixed point was the bed temperature that was controlled by changing the fuel feeding in parallel with adjustment of the FGR and air (in semi-oxyfuel cases) in order to keep the oxygen concentration in the flue gas constant. The third fixed point was the pressure drop across the bed controlled together with the bed temperature. Results of experiments and calculations are summarized in tables 3 (semi-oxyfuel) and 4 (full oxyfuel and air-only reference). All measured values presented in the tables are mean values of the variables over the entire stable period of the specific case. The stable operation was in all cases at least 1 hour.

In the table 4, the mark *) indicates that the oxyfuel mode was not completely full. A certain flow of primary air that was necessary to maintain the pressure balance in the oxy BFB rig. This residual flow could not be avoided due to the current set-up of the rig. These residual 2 m^3_N/h of air however cause the CO_2 concentration to reach only approx. 75 % in dry flue gas. Even if the air intake is a relatively small fraction (about 3.5 %) in comparison with the volumetric flows of the FGR and the oxygen, its contribution makes still about 19 vol. % of nitrogen in the outgoing flue gas. The data in both tables show a good agreement of calculation results and experiments and generally prove that the presented approach is applicable for oxyfuel calculations. The relative difference between measured and calculated values does not exceed 10 % which can be considered as satisfactory. There are however several remarkable points. First, one should notice that the measured air flows and fluidization velocities are always higher than the calculated. In parallel, the measured and calculated CO_2 concentrations are in a good agreement. This indicates that some of the gas stream is lost in the rig due to leakages. With high probability this is caused by the overpressure in the combustor. The modularity of the rig involves a number of connecting flanges that need to be properly sealed, which is of course complicated. According to the results, about 10 % of the stream flow into the rig can be lost by leakages.

Table 3. Results, semi-oxyfuel mode; O_2 and CO_2 concentrations in dry gas

	Semi-oxyfuel 1		Semi-oxyfuel 2		Semi-oxyfuel 3		Semi-oxyfuel 4	
Parameter	Measured	Calculated	Measured	Calculated	Measured	Calculated	Measured	Calculated
Bed temperature (°C)	892		892		890		889	
Fuel feeding (kg/h)	5.08		4.82		4.86		5.11	
Power load (kW)	30		28		29		30	
FGR temperature (°C)	46.6		49.6		52.0		54.9	
Oxygen (m^3_N/h)	1.5		2		2.5		3	
Air (m^3_N/h)	21.2	20.0	21.0	17.3	18.0	15.4	17.5	15.0
FGR (m^3_N/h)	24.1	24.1	25.5	25.6	29.3	29.3	31.1	31.1
r (-)	0.89		1.03		1.24		1.27	
T-AD (°C)	1441		1406		1362		1355	
O_2 flue gas (%)	2.41	2.41	2.56	2.56	3.19	3.19	3.81	3.81
CO_2 flue gas (%)	21.83	21.75	24.26	23.17	26.50	25.22	27.59	26.50
O_2 in fluid. stream (%)	15.17		15.22		15.68		16.74	
uf (m/s)	1.64	1.48	1.70	1.44	1.74	1.49	1.81	1.54

Table 4. Results, full oxyfuel and air-only reference; O_2 and CO_2 concentrations in dry gas

	Full oxyfuel		Air reference
Parameter	Measured	Calculated	Calculation only
Bed temperature (°C)	895		900
Fuel feeding (kg/h)	5.98		5.40
Power load (kW)	35		27.5
FGR temperature (°C)	71.4		50
Oxygen (m^3_N/h)	6.7		0
Air (m^3_N/h)	2.00 *)	1.13	31.8
FGR (m^3_N/h)	45.7	45.8	11.4
r (-)	4.95		0.30
T-AD (°C)	1235		1479
O_2 flue gas (%)	5.77	5.77	4.92
CO_2 flue gas (%)	75.34	73.86	14.55
O_2 in fluid. stream (%)	19.50		17.30
uf (m/s)	1.91	1.66	2.05

The second noteworthy point is the T-AD. As expected, the T-AD decreases with increasing degree of the oxyfuel mode and is the lowest for full oxyfuel. This decrease is caused by the rising specific heat capacity of the gas stream while the volumetric flow is maintained at roughly constant value. The balance to keep the fluidized bed temperature constant must be ensured by increasing of the fuel feeding, i.e. by an increased power load. This situation is shown in figure 3.

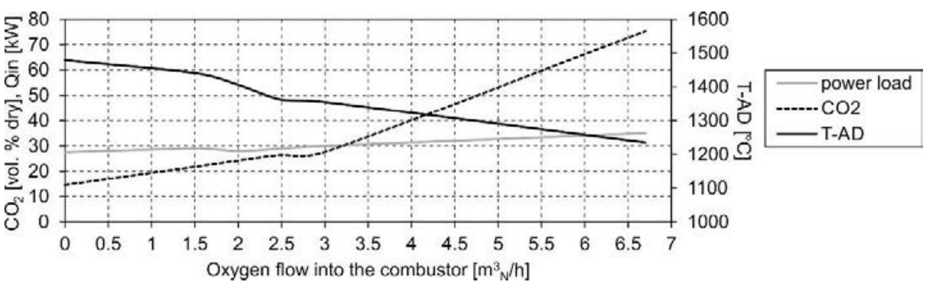

Fig. 3. Changes of T-AD, input power load and CO_2 concentration with increasing flow of oxygen into the BFBC

It is also worth to comment on fluidization velocities in the experiments. When comparing the results with table 2, it becomes obvious that the fluidization velocity is at about 10-12 multiple of the minimum fluidization velocity and that it is nearly at the terminal particle velocity. This shows a well-developed fluidization in the dense bed zone. The measured fluidization velocity is appropriately larger than the calculated value, since it is obtained from the sum of all flows entering the combustor.

5. Conclusion

The calculation procedure for oxyfuel combustion in a BFB was verified by several experiments in semi-oxyfuel and full oxyfuel conditions. The major variable for comparison of the calculated and measured results is CO_2 concentration that agrees within less than 10 % relative difference. This can be considered as satisfactory. To keep the fluidization regime similar in all cases, it is necessary to use the FGR to sustain the flow of primary stream at a constant level. The FGR introduces more CO_2 with a higher specific heat capacity and thus reduces the adiabatic flame temperature. It was shown that from air-only mode to full oxyfuel mode the adiabatic flame temperature drops by about 250°C. In order to keep the bed temperature constant along all tested cases from air only to full oxyfuel, it was essential to increase the power input to the combustor or in other words to increase the volumetric load of the fluidized bed. The decrease of the T-AD calculated to 14 % (from 1752 to 1508 K) corresponds to increase of the power input by 20 % (from 28 to 35 kW), which can be also considered as a satisfactory agreement. The power increase corresponds to an increase of the volumetric load in the combustor from 3.3 to 4.1 MW/m^3.

Acknowledgment

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